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(54) ORGANIC ELECTROLUMINESCENT ELEMENT

(57)Abstract:
PROBLEM TO BE SOLVED: To enhance light taking out efficiency by forming a region, where the turbulence of reflection and angle of refraction of light on the interface is produced, in at least one in the vicinity of the interface of an organic layer and an electrode layer in a pair of electrode layers between which the organic layer of a luminescent layer is interposed.
SOLUTION: An organic electroluminescent element is constituted by interposing an organic layer containing a luminescent layer between a pair of electrode layers whose at least one is transparent. A region which is formed by heterogeneously dispersing two or more kinds of materials and where the turbulence of reflection and angle of refraction of light is produced is formed in at least one in the vicinity of the boundary between at least one electrode layer and the organic layer. Irregular reflection and irregular refraction of light are produced, trap of light in the layer caused by total reflection is reduced, and the taking out efficiency of light is enhanced. The interface is smooth, and local, uneven electric characteristics are not generated, and uneven luminescence and dielectric breakdown are not generated.

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CLAIMS

[Claim(s)]

[Claim 1] In the organic electroluminescent element of the structure which pinched the organic layer of one side [at least] containing a luminous layer in the electrode layer of a transparent pair The organic layer near the interface at least with the organic layer of one [at least] electrode layer, The organic electroluminescent element characterized by preparing the field which makes reflection and angle of refraction of the light in the above-mentioned interface which distributed two or more sorts of ingredients to the ununiformity, and was formed at least in one side in near the interface with one [at least] electrode layer produce turbulence.

[Claim 2] The organic electroluminescent element characterized by preparing the field which the front face by the side of the ejection of light is made [field] into a scattered reflection side among the components concerned, or makes reflection and angle of refraction of the light in the above-mentioned front face produce turbulence in the organic electroluminescent element equipped with the organic layer containing a luminous layer near the front face by the side of the ejection of the above-mentioned light.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to an organic electroluminescent element.

[0002]

[Description of the Prior Art] The hole and electron which were poured in from the electrode layer of the pair which sandwiches a luminous layer recombine luminescence of an organic electroluminescent element within the above-mentioned luminous layer, it generates an exciton, and is considered to be based on exciting the molecule of luminescent material with which it constitutes a luminous layer. And if a fluorochrome is used as a luminescent material, an emission spectrum equivalent to the photoluminescence of the coloring matter molecule concerned will be obtained as electroluminescence luminescence.

[0003] [C.W.Tang and S.A.VanSlyke which carries out green luminescence efficiently

by the low battery about 10V, compared with the conventional organic electroluminescent element recently equipped with the luminous layer of monolayer structure and as which the component equipped with two-layer [of a hole transportation layer and an electronic transportability luminous layer] was proposed by Tang and Vanslyke; Appl.Phys.Lett., 51 (1987) 913]. The configurations of a component are the anode plate formed on the glass substrate, a hole transportation layer, an electronic transportability luminous layer, and cathode.

[0004] With the above-mentioned component, while a hole transportation layer serves to pour in a hole from an anode plate to an electronic transportability luminous layer, it prevented escaping to an anode plate, without the electron poured in from cathode recombining with a hole, and the role which confines an electron into an electronic transportability luminous layer is also played. For this reason, according to the containment effectiveness of the electron by this hole transportation layer, compared with the component of the conventional monolayer structure, the recombination of a hole and an electron happens more efficiently, and the sharp fall of driver voltage is attained.

[0005] Moreover, Saito and others sets for the component of two-layer structure. It was shown that not only an electronic transportation layer but a hole transportation layer can turn into a luminous layer (hole transportability luminous layer), and also they are [C.Adachi and T.Tsutsui and S.Saito.; Appl.Phys.Lett., 55 (1989) 1489], Between a hole transportation layer and an electronic transportation layer [C.Adachi which proposed the organic electroluminescent element of the three-tiered structure into which the luminous layer was inserted, S.Tokito, T.Tsutsui and S.Saito; Jpn.J.Appl.Phys., 27 (1988) L269].

[0006] The component of Saito's and others two-layer structure consisted of the anode plate formed on the glass substrate, a hole transportability luminous layer, an electronic transportation layer, and cathode, and it prevented escaping to cathode, without the hole poured into a previous thing and previous reverse from the anode plate while the electronic transportation layer served to pour in an electron from cathode to a hole transportability luminous layer recombining with an electron, and the role which confines a hole into a hole transportability luminous layer has also been played. For this reason, the sharp fall of driver voltage is attained like a previous thing according to the containment effectiveness of the hole by this electronic transportation layer.

[0007] Moreover, the component of Saito's and others three-tiered structure is what improved previous Tang's and others component further. While serving to consist of

the anode plate formed on the glass substrate, a hole transportation layer, a luminous layer, an electronic transportation layer, and cathode, and for a hole transportation layer to confine an electron in a luminous layer In order that an electronic transportation layer may serve to confine a hole in a luminous layer, compared with the thing of two-layer structure, the recombination effectiveness of the electron within a luminous layer and a hole improves further. Moreover, the above-mentioned electronic transportation layer and a hole transportation layer serve to prevent escaping and carrying out quenching of an electron and the exciton generated by the recombination of a hole to the electrode layer of one of yin and yang. Therefore, according to the component of the three-tiered structure which Saito and others proposed, luminous efficiency improves further.

[0008]

[Problem(s) to be Solved by the Invention] External energy-efficiency etaE which shows the luminous efficiency of an electroluminescent element according to Tsutsui (ext) Internal energy effectiveness etaE of a component (int) And ejection effectiveness etaEXT of light Formula (a) : [0009]

[Equation 1]

$$\eta_E(\mathrm{ext}) = \eta_{EXT}\,\eta_E(\mathrm{int}) \qquad \text{(a)}$$

[0010] ["latest advance of organic electroluminescence" Tetsuo Tsutsui in ******, display , and imaging 1994, vol.3, pp.237-245]. Moreover, it is internal energy effectiveness etaE (int) of a component among the above. Formula (b) : [0011]

[Equation 2]

$$\eta_E(\mathrm{int}) = \eta_\phi(\mathrm{int})\,\frac{\varepsilon_P}{V} \qquad \text{(b)}$$

[0012] **** profit and internal-quantum-efficiency etaphi (int) Average energy epsilonP of applied voltage V and the photon emitted (eV) asks and internal-quantum-efficiency etaphi (int) is a formula (c) further. : [0013]

[Equation 3]

$$\eta_\phi(\mathrm{int}) = \gamma\,\eta_r\,\eta_f \qquad \text{(c)}$$

[0014] Generation effectiveness etar of **** profit, an electron, and the impregnation balance factor gamma of a hole and the singlet exciton by carrier recombination Luminescence quantum efficiency etaf from a singlet exciton It asks. And it is internal-energy effectiveness etaE (int) of a component for the purpose of

improvement in the luminous efficiency of the former and an electroluminescent element. It is generation effectiveness etar of the above-mentioned impregnation balance factor gamma and a singlet exciton in order to raise. And luminescence quantum efficiency etaf from a singlet exciton About improvement, examination is made variously.

[0015] For example, although the impregnation balance factor gamma is a multiplier which shows the impregnation balance of an electron, the electron with which a hole sets to 1.0 equivalence and the ideal condition of being poured in, and a hole and it is usually gamma< 1.0, in the case of an organic electroluminescent element, it can bring close to 1.0 by improving the balance of the impregnation of an organic layer as multilayer structure more than two-layer with an electron and a hole as mentioned above, for example.

[0016] Moreover, if an idea that a triplet exciton and a singlet exciton occur at a rate of 3:1 is followed by the difference in the spin multiplicity at the time of an electron and a hole recombining, it will be generation effectiveness etar of a singlet exciton. Maximum is above-mentioned generation effectiveness etar, if it even takes into consideration that the generated triplet exciton converts into a singlet exciton, although it is 0.25. It increases to 0.40.

[0017] Furthermore, it is luminescence quantum efficiency etaf from a singlet exciton. If luminescence from one molecule which was mainly reflecting the property of photogene propers, such as a fluorochrome, and was isolated simply is taken into consideration, it will be fluorescence quantum efficiency [of a proper]: [0018] to a photogene molecule or a molecule solid-state.

[Equation 4]

Φ_{ϕ}

[0019] Although it is equal, in an actual component, there is loss of the ******** energy by various kinds of energy processes accompanying the lamination of a component. Then, the lamination of a component is devised and it is luminescence quantum efficiency etaf from a singlet exciton to make an organic layer into the multilayer structure more than two-layer as mentioned above, while enlarging fluorescence quantum efficiency of the molecule itself and the molecule solid-state itself etc. Bringing close to 1.0 is considered.

[0020] ejection effectiveness etaEXT of the light which is another factor which determines the luminous efficiency of an electroluminescent element on the other hand **** -- the optical structure of a component is involved. Namely, although it is

necessary to pass the interface (for the interface of the outermost layer of a component and atmospheric air to be included) of the medium by which some refractive indexes differ in order to emit luminescence by the luminous layer into atmospheric air For the light which carried out incidence to the field side the include angle beyond the critical angle when following the law of refraction of Snell, since total reflection is carried out by the interface, it is confined into a layer, the inside of the layer concerned is guided and it disappears, only the part is ejection effectiveness etaEXT of light. It falls.

[0021] Ejection effectiveness etaEXT of light in the case of taking out field-like luminescence generated in an electroluminescent element in the medium of the refractive index n (n> 1.0) which constitutes a luminous layer in the atmospheric air whose refractive index is 1.0 When the total reflection in the interface mentioned above is taken into consideration, they are the above-mentioned refractive index n and the following formula (d). : [0022]

[Equation 5]

$$\eta_{EXT} \sim \frac{1}{2n^2} \qquad \text{(d)}$$

[0023] Coming out and having the relation expressed is known. If this is applied to an organic electroluminescent element, since the refractive index n of the organic material which constitutes organic layers, such as a luminous layer, is about 1.6 Ejection effectiveness etaEXT of the light in an organic electroluminescent element Although about 20% of the light which was set to about 0.195 and generated in the luminous layer is taken out out of a component As for about 80% of remainder, it turns out that it is confined into a component by the total reflection in the interface of each class which constitutes a component as mentioned above, and the inside of a layer is guided and it disappears.

[0024] for this reason, internal energy effectiveness etaE (int) mentioned above Improvement, simultaneously ejection effectiveness etaEXT of light Raising is an important technical problem towards utilization of a future organic electroluminescent element. For example, Inoguchi makes the interface of each class which constitutes a component a scattered reflection side that the containment to the inside of the layer by the total reflection in the interface of the light which emitted light by the luminous layer should be reduced, or has proposed distributing the particle which takes the dispersion lead in a luminous layer ["electroluminescent display" Toshio Inoguchi, the Sangyo Tosho Publishing **********, and July 25, Heisei 3 first-edition issue].

[0025] However, Inoguchi has pointed out in this proposal also about the trouble caused by the two above-mentioned law. That is, if it takes into consideration that the high voltage is the need, that the thickness of each class which constitutes a component is thin, etc. to the drive of an electroluminescent element in order that electric field may concentrate on a height among the minute irregularity which constitutes this scattered reflection side, when making the interface of a layer into a scattered reflection side, it is supposed that there is a possibility that dielectric breakdown of a component may arise, from the above-mentioned height.

[0026] Moreover, in the luminous layer, when distributing the particle which takes the dispersion lead and which has the grain boundary which progressed clearly, by the particle concerned, the local ununiformity was produced in the electrical characteristics of a luminous layer, and it has burned [there is a possibility that the ununiformity and dielectric breakdown of luminescence by it may arise, and]. The purpose of this invention is to offer the new organic electroluminescent element which raised the ejection effectiveness of light, without producing an ununiformity, dielectric breakdown, etc. of luminescence.

[0027]

[Means for Solving the Problem] The 1st organic electroluminescent element of this invention for solving the above-mentioned technical problem It is the organic electroluminescent element of the structure which pinched the organic layer of one side [at least] containing a luminous layer in the electrode layer of a transparent pair. The organic layer near the interface at least with the organic layer of one [at least] electrode layer, It is characterized by preparing the field which makes reflection and angle of refraction of the light in the above-mentioned interface which distributed two or more sorts of ingredients to the ununiformity, and was formed at least in one side in near the interface with one [at least] electrode layer produce turbulence.

[0028] In the 1st organic electroluminescent element of this invention of this, scattered reflection and the containment to the inside of the layer by the total reflection of light since it is random-refracted decrease [light] by the field which makes reflection and angle of refraction of the light in the above-mentioned interface established near the interface of an organic layer and an electrode layer produce turbulence, and the ejection effectiveness of light improves. Moreover, the above-mentioned interface is smooth, and although the field which was prepared near [this] the interface when it did not have irregularity with a possibility that electric field may concentrate and which makes reflection and angle of refraction of the light in the interface concerned produce turbulence distributes two or more sorts of

ingredients, both do not have a grain boundary which produces a local ununiformity in the electrical characteristics of a layer and which progressed clearly.

[0029] Therefore, according to the 1st organic electroluminescent element of this invention, it becomes possible to raise the ejection effectiveness of light, without producing an ununiformity, dielectric breakdown, etc. of luminescence. Moreover, the 2nd organic electroluminescent element of this invention It is the organic electroluminescent element equipped with the organic layer containing a luminous layer. It is characterized by preparing the field which makes reflection and angle of refraction of the light in the above-mentioned front face which made the front face by the side of the ejection of light the scattered reflection side among the components concerned, or distributed two or more sorts of ingredients to the ununiformity, and was formed near the front face by the side of the ejection of the above-mentioned light produce turbulence.

[0030] the account of a top -- the 2nd organic electroluminescent element of this invention The front face by the side of the ejection of the light of a component which is not related to the electrical characteristics of a luminous layer [whether it considers as a scattered reflection side as mentioned above, and] Or without producing an ununiformity, dielectric breakdown, etc. of luminescence near [front face] the above because of what prepared the field which makes reflection and angle of refraction of the light in the front face concerned produce turbulence, the containment to the inside of the layer by the total reflection of light can be reduced, and the ejection effectiveness of light can be raised.

[0031] In addition, the field which makes reflection and angle of refraction of a light here produce turbulence is a field which has the function to make either whenever [in the interface or front face of the light which emitted light by the luminous layer among organic layers, and carried out incidence to the interface with an electrode layer, or the front face by the side of the ejection of the light of a component concerned / angle-of-reflection], or whenever [angle-of-refraction], and both produce turbulence.

[0032]

[Embodiment of the Invention] First, the 1st organic electroluminescent element of this invention is explained. The organic electroluminescent element of the above 1st has the structure which pinched the organic layer of one side [at least] which contains a luminous layer at least in the electrode layer of a transparent pair.

[0033] An organic layer may be the monolayer structure of having only a luminous layer, as usual, and may be multilayer structure, such as two-layer [which was

combined with various layers as the luminous layer was mentioned above] or three layers, and four etc. layers. the combination of the hole transportability luminous layer and electronic transportation layer which were mentioned above as an example of two-layer structure, for example -- or the combination of an electronic transportability luminous layer and a hole transportation layer etc. is raised.

[0034] Moreover, a hole transportability luminous layer besides the combination of the hole transportation layer, luminous layer, and electronic transportation layer which were mentioned above as an example of a three-tiered structure, for example, The hole block layer which serves to prevent escaping to cathode, without the hole poured in from the anode plate recombining with an electron, and to confine a hole into a hole transportability luminous layer, combination with an electronic transportation layer -- or it prevents escaping to an anode plate, without an electronic transportability luminous layer and the electron poured in from cathode recombining with a hole, and the combination of the electronic block layer which serves to confine an electron into an electronic transportability luminous layer, and a hole transportation layer etc. is raised.

[0035] the combination of the hole impregnation layer which serves to rescue that a hole is injected into a hole transportation layer, for example from an anode plate as an example of further 4 layer structures, a hole transportation layer, a luminous layer, and an electronic transportation layer -- or the combination of the above-mentioned hole impregnation layer, a hole transportability luminous layer, a hole block layer, and an electronic transportation layer etc. is raised. The above-mentioned organic layer is combined with the yin-and-yang two-electrodes layer for injecting an electron and a hole into the organic layer concerned. Under the present circumstances, in order to take out luminescence from a luminous layer out of a component, at least one side of the above-mentioned two-electrodes layers is formed with transparence electrical conducting materials, such as ITO (indium tin oxide), as mentioned above.

[0036] If protection of a component is taken into consideration, above-mentioned each class forms a laminating and the base material formed with glass, transparence plastic film, etc., it is desirable to make the base material side concerned into the ejection side of the light from a luminous layer, and it should just form the electrode layer of the side formed in right above [of a base material] with the above-mentioned transparence electrical conducting material in this case. Moreover, on the electrode layer of the side formed on an organic layer, that a component should be protected from moisture, oxygen, etc. in air, a protective layer may be formed or the component in which the protective layer was formed may be further closed by glass or the

polymer.

[0037] In this invention, the field which makes the reflection and angle of refraction of light which distributed two or more sorts of ingredients to the ununiformity at least near the interface with one [at least] electrode layer of the above-mentioned organic layer near the interface with the organic layer of one [at least] electrode layer as mentioned above among the organic electroluminescent elements of the above-mentioned class structure produce turbulence is prepared.

[0038] Specifically, it is (1). The whole electrode layer near the interface with the organic layer of one side of the electrode layer of the pair whose organic layer is pinched, or both, and (2) (3) The field which makes it produce turbulence in reflection and angle of refraction of light whether they are both the above (1), (2), and ******** is formed. [one side of the electrode layer of the pair among organic layers, or near / both / the interface]

[0039] What is necessary is to be able to consider various approaches, in order to establish the above-mentioned field in a predetermined part, but to form by coincidence vacuum evaporationo of two or more sorts of mutually different ingredients, or just to dope the ingredient of another side in the layer which formed the layer concerned with one ingredient, in order to establish the above-mentioned field in the layer formed, for example of vapor growth, such as a vacuum deposition method. For example, what is necessary is just to form the electrode layer concerned like the example mentioned later by the with a vacuum evaporationo velocity ratio of about 10:1 vapor codeposition of magnesium (Mg) and silver (Ag), in order to prepare the field which makes the electrode layer of the side which is not transparent produce turbulence in reflection and angle of refraction of light, and the field which makes the angle of reflection of light produce turbulence in more detail.

[0040] Thus, as a result of becoming the structure distributed without having the grain boundary where a MgAg alloy phase is clear in the continuous phase of Mg and producing turbulence in the angle of reflection of the light from a luminous layer as mentioned above, the formed electrode layer reduces the containment to the inside of the layer by the total reflection of light, and raises the ejection effectiveness of light. In addition, in the above-mentioned electrode layer, two of the followings can be considered, for example as a cause which produces turbulence in the angle of reflection of the light from a luminous layer.

** It has various (since a bi-phase does not have the grain boundary which progressed clearly as mentioned above) phases, such as Mg phase, a MgAg phase, and both intermediate phase, since the interactions to the incident light of each phase

differ, a difference arises in angle of reflection, consequently the interface with the organic layer of an electrode layer produces turbulence in the angle of reflection of the light from a luminous layer.

** By the oxygen contained in an ambient atmosphere or an organic layer in the case of component production, since it changes with each above-mentioned phases from which the interface of the above-mentioned electrode layer and an organic layer has oxidized very slightly, and the presentation of the oxidizing zone constitutes an electrode layer, a difference arises also to the rate of optical refraction, consequently produce turbulence in the angle of reflection of the light from a luminous layer.

[0041] In addition, what is necessary is just to distribute at an ununiformity the transparent material from which two or more sorts of refractive indexes differ in transparent layers other than the above-mentioned electrode layer, in order to form in reflection and angle of refraction of light the field which produces turbulence. In the decentralized structure of two or more sorts of ingredients which make reflection and angle of refraction of light produce turbulence, although especially the magnitude (for example, magnitude of the MgAg alloy phase distributed in the continuous phase of Above Mg etc.) of the above-mentioned decentralized structure is not limited, it is desirable that they are 1 of the luminescence wavelength from a luminous layer / about 10 to 10 times.

[0042] When the magnitude of a decentralized structure is under the above-mentioned range or exceeds the above-mentioned range, there is a possibility that the effectiveness of reducing the total reflection in the interface of luminescence from a luminous layer may become inadequate by turbulence of reflection and angle of refraction. In addition, as for especially the magnitude of the above-mentioned decentralized structure, it is desirable that above-mentioned within the limits is also luminescence wavelength and below actual size much more.

[0043] Below, the 2nd organic electroluminescent element of this invention is explained. The organic electroluminescent element of the above 2nd is equipped with the organic layer containing a luminous layer. Monolayer structure or the laminated structure more than two-layer is sufficient as the lamination of an organic layer like the case of the 1st previous organic electroluminescent element.

[0044] An electrode layer and a base material as well as the case of the 1st previous organic electroluminescent element can constitute. Namely, if protection of a component is taken into consideration, an organic layer and an organic electrode layer should just form the electrode layer of the side desirable [as transparence base materials, such as glass and transparence plastic film, / the base material side

concerned / into the ejection side of the light from a luminous layer] and formed [a laminating and the base material formed] in right above [of a base material] in this case with a transparence electrical conducting material.

[0045] Moreover, on the electrode layer of the side formed on an organic layer, that a component should be protected from moisture, oxygen, etc. in air, a protective layer may be formed or the component in which the protective layer was formed may be further closed by glass or the polymer. In this invention, the field which makes the reflection and angle of refraction of light which made the front face by the side of the ejection of light the scattered reflection side, or distributed two or more sorts of ingredients to the ununiformity, and was formed near the front face by the side of the ejection of the above-mentioned light as mentioned above among the organic electroluminescent elements of the above-mentioned class structure produce turbulence is prepared.

[0046] When the ejection side of ** light is a transparence base material side as mentioned above, specifically The front-face [in which each class of the transparence base material concerned is formed / of a side and the opposite side], and ejection side of ** light When it is the electrode layer side of a base material and the opposite side and a protective layer is formed in the front face of the electrode layer concerned, and the front face of the ** above-mentioned electrode layer, the front face of the protective layer concerned and the field which it is made [field] into a scattered reflection side whether to be ********, or makes reflection and angle of refraction of light produce turbulence near each above-mentioned front face are formed.

[0047] In addition, as for the transparence base material of **, it is [among these] desirable to roughen the front face by approaches, such as mechanical polishing, and to consider as a scattered reflection side in respect of workability etc. It is forming by coincidence vacuum evaporationo of two or more sorts of ingredients which are mutually different as mentioned above because of that in which the electrode layer of ** and the protective layer of ** are formed mainly with vacuum deposition on the other hand, or doping the ingredient of another side in the layer formed with one ingredient, and it is desirable to form the field which makes reflection and angle of refraction of light produce turbulence near the front face.

[0048] Although especially the magnitude of the irregularity which constitutes the above-mentioned scattered reflection side is not limited, it is desirable that the RMS values showing extent of the irregularity of a scattered reflection side are 1 of the luminescence wavelength from a luminous layer / about 10 to 10 times. When an RMS

value is under the above-mentioned range or exceeds the above-mentioned range, there is a possibility that the effectiveness of reducing the total reflection in the interface of luminescence from a luminous layer may become inadequate by scattered reflection.

[0049] In addition, as for especially the above-mentioned RMS value, it is desirable that above-mentioned within the limits is also luminescence wavelength and below actual size much more. As for the magnitude of the decentralized structure of two or more sorts of ingredients which, on the other hand, constitute the field which makes the reflective angle of refraction of light produce turbulence, it is desirable that they are that explanation of the 1st previous organic electroluminescent element described and a homotype enclosure.

[0050] In the organic electroluminescent element of the above 2nd, the configuration of the 1st organic electroluminescent element mentioned above may be introduced into one [at least] interface among a further organic layer and the layer of the pair which sandwiches it. That is, near [interface] the above, the field which makes the reflection and angle of refraction of light which distributed two or more sorts of ingredients to the ununiformity, and was formed produce turbulence may be prepared. In this case, the reduction effectiveness of the total reflection in the front face by the side of the ejection of light and the reduction effectiveness of the total reflection in the above-mentioned interface can improve the luminous efficiency of a component further conjointly.

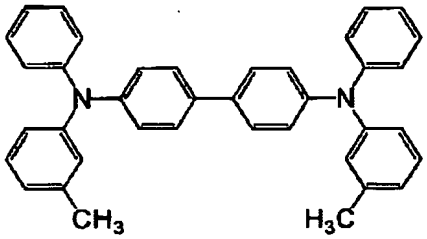
[0051]

[Example] This invention is explained based on an example and the example of a comparison below.

ITO (indium tin oxide) was vapor-deposited with example 1 electron-beam vacuum deposition, it was made to wash and dry in a boiling methanol, and the glass base material in which the thickness of 150-160nm and the ITO electrode layer of 15ohms of sheet resistance and ** were formed was further defecated in UV ozone ashing, after using a detergent (SEMIKO -- clean), distilled water, an acetone, and isopropanol for this order and cleaning them ultrasonically.

[0052] It is a formula (1) first on the above-mentioned ITO electrode layer by the bottom of the condition of ultimate-vacuum:1x10-6Torr, a substrate temperature:room temperature, an evaporation rate 0.2 - 0.3 nm/s, and the vacuum deposition method in the condition of having set this glass base material in the vacuum evaporation system next, and having equipped with the mask corresponding to the configuration of a luminescence field. : [0053]

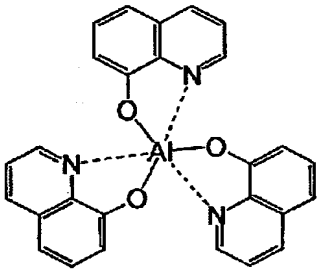
[Formula 1]



(1)

[0054] It comes out, the N [which is expressed], N'-diphenyl-N, and N'-screw (3-methylphenyl) -1, the 1'-biphenyl -4, and 4-diamine are vapor-deposited, the hole transportation layer of 40nm of thickness is formed, and then it is a formula (2) by the vacuum deposition method at these conditions succeedingly on this hole transportation layer. : [0055]

[Formula 2]



(2)

[0056] Tris (8-quinolate) aluminum come out of and expressed (III) The complex was vapor-deposited and the electronic transportability luminous layer of 60nm of thickness was formed. Ultimate-vacuum:1x10-6Torr, substrate temperature after exchanging a mask for the thing corresponding to the configuration of cathode in atmospheric air next : with a vacuum deposition method under a room temperature and the condition of evaporation rate 1 nm/s Vapor codeposition of Mg and Ag is carried out by the evaporation rate ratio 10:1 on the above-mentioned electronic transportation layer. After forming Mg/Ag=10 / 1 (mole ratio), and the Mg/Ag electrode layer of 150nm of thickness, on it, the independent vacuum evaporationo of Ag was carried out, the protective layer of 100nm of thickness was formed, and the organic electroluminescent element was manufactured.

[0057] And when direct current voltage was impressed between two electrodes in a room temperature and atmospheric air, having used the anode plate and the Mg/Ag

electrode layer as cathode for the ITO electrode layer of the above-mentioned organic electroluminescent element, green luminescence (luminescence peak 53nm) from an electronic transportability luminous layer was observed on the electrical potential difference beyond 4V. moreover, the place which measured the luminescence brightness of the above-mentioned component using the luminance meter [LS-100 by Minolta Co., Ltd.] -- the highest brightness -- 20000 cd/m2 it is -- moreover, when the luminous efficiency of a component was searched for from this result, the highest luminous efficiency was 1.5 lm/W.

[0058] Furthermore, when X diffraction measurement of the above-mentioned Mg/Ag electrode layer was carried out, since the peak of a MgAg alloy was detected in addition to the peak of Mg as this Mg/Ag electrode layer was shown in drawing 1 , it turned out that it has the structure where the MgAg alloy phase was distributed, in the continuous phase of Mg. And observation by the metaloscope of the above-mentioned Mg/Ag electrode layer showed that the above-mentioned MgAg alloy phase was distributed in the condition of not having a clear grain boundary in the continuous phase of Mg.

It replaced with the example of comparison 1 Mg/Ag electrode layer, and the organic electroluminescent element was manufactured like the example 1 except having formed the independent vacuum evaporationo layer (150nm of thickness) of Mg.

[0059] And when direct current voltage was impressed between two electrodes in a room temperature and atmospheric air, having used the anode plate and the Mg/Ag electrode layer as cathode for the ITO electrode layer of the above-mentioned organic electroluminescent element, green luminescence (luminescence peak 53nm) from an electronic transportability luminous layer was observed on the electrical potential difference beyond 4V. moreover, the place which measured the luminescence brightness of the above-mentioned component using the luminance meter [LS-100 by Minolta Co., Ltd.] -- the highest brightness -- 8000 cd/m2 it is -- moreover, when the luminous efficiency of a component was searched for from this result, the highest luminous efficiency was 1.0 lm/W.

The organic electroluminescent element was manufactured like the example 1 except having carried out mechanical polishing of the front face of the side which forms the ITO electrode layer of the glass base material equivalent to the front face by the side of the ejection of light of two examples, and the opposite side so that surface roughness might be set to 10-20nm with an RMS value.

[0060] And when direct current voltage was impressed between two electrodes in a room temperature and atmospheric air, having used the anode plate and the Mg/Ag

electrode layer as cathode for the ITO electrode layer of the above-mentioned organic electroluminescent element, green luminescence (luminescence peak 53nm) from an electronic transportability luminous layer was observed on the electrical potential difference beyond 4V. moreover, the place which measured the luminescence brightness of the above-mentioned component using the luminance meter [LS-100 by Minolta Co., Ltd.] -- the highest brightness -- 43000 cd/m2 it is -- moreover, when the luminous efficiency of a component was searched for from this result, the highest luminous efficiency was 3.81 m/W.

It replaced with the example 3 Mg/Ag electrode layer, and the organic electroluminescent element was manufactured like the example 2 except having formed the aluminum/Li electrode layer of 150nm of thickness, and the 0.05 % of the weight of the degrees of Li ** with the vacuum deposition method using the aluminum (aluminum) lithium (Li) alloy (the 2 % of the weight of the degrees of Li **) as evaporation matter.

[0061] And when direct current voltage was impressed between two electrodes in a room temperature and atmospheric air, having used the anode plate and the aluminum/Li electrode layer as cathode for the ITO electrode layer of the above-mentioned organic electroluminescent element, green luminescence (luminescence peak 53nm) from an electronic transportability luminous layer was observed on the electrical potential difference beyond 4V. moreover, the place which measured the luminescence brightness of the above-mentioned component using the luminance meter [LS-100 by Minolta Co., Ltd.] -- the highest brightness -- 21000 cd/m2 it is -- moreover, when the luminous efficiency of a component was searched for from this result, the highest luminous efficiency was 1.6 lm/W.

[0062] In addition, when the above-mentioned aluminum/Li electrode layer was observed with the metaloscope, it turned out that it is the uniform continuation layer of an AlLi alloy.

[0063]

[Effect of the Invention] As explained in full detail, as mentioned above, the organic electroluminescent element of this invention Near the front face by the side of the ejection of the light of a component near the interface of an organic layer and an electrode layer It becomes possible to raise the ejection effectiveness of light, without producing an ununiformity, dielectric breakdown, etc. of luminescence because of what prepared the field which makes the reflection and angle of refraction of light which distributed two or more sorts of ingredients to the ununiformity, and was formed produce turbulence, or made the front face by the side of the ejection of the

light of a component the scattered reflection side.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the graph in the organic electroluminescent element manufactured in the example 1 of this invention which shows the X diffraction spectrum of a MgAg electrode layer.

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(54)【発明の名称】　有機エレクトロルミネッセンス素子

(57)【要約】

【課題】　発光の不均一や絶縁破壊等を生じることなく、光の取り出し効率を向上させた、新規な有機エレクトロルミネッセンス素子を提供する。

【解決手段】　有機の層と電極層との界面近傍、または素子の光の取り出し側の表面近傍に、２種以上の材料を不均一に分散して形成された、光の反射・屈折角に乱れを生じさせる領域を設けるか、または素子の光の取り出し側の表面を乱反射面とした。

【特許請求の範囲】
【請求項１】発光層を含む有機の層を、少なくとも一方が透明である一対の電極層で挟んだ構造の有機エレクトロルミネッセンス素子において、少なくとも一方の電極層の、少なくとも有機の層との界面近傍、ならびに有機の層の、少なくとも一方の電極層との界面近傍のうち少なくとも一方に、２種以上の材料を不均一に分散して形成された、上記界面での光の反射・屈折角に乱れを生じさせる領域を設けたことを特徴とする有機エレクトロルミネッセンス素子。
【請求項２】発光層を含む有機の層を備えた有機エレクトロルミネッセンス素子において、当該素子のうち、光の取り出し側の表面を乱反射面とするか、または上記光の取り出し側の表面近傍に、上記表面での光の反射・屈折角に乱れを生じさせる領域を設けたことを特徴とする有機エレクトロルミネッセンス素子。
【発明の詳細な説明】
【０００１】
【発明の属する技術分野】この発明は、有機エレクトロルミネッセンス素子に関するものである。
【０００２】
【従来の技術】有機エレクトロルミネッセンス素子の発光は、発光層を挟む一対の電極層から注入されたホールと電子とが上記発光層内で再結合して励起子を生成し、それが発光層を構成する発光材料の分子を励起することに基づくと考えられている。そして、発光材料として蛍光色素を使用すると、当該色素分子のフォトルミネッセンスと同等の発光スペクトルが、エレクトロルミネッセンス発光として得られる。
【０００３】近時、単層構造の発光層を備えた従来の有機エレクトロルミネッセンス素子に比べて、約１０Ｖという低電圧で効率よく緑色発光する、ホール輸送層と電子輸送性発光層の２層を備えた素子が、TangとVanslykeによって提案された〔C. W.Tang and S. A. VanSlyke; Appl. Phys. Lett. , 51 (1987) 913〕。素子の構成は、ガラス基板上に形成した陽極、ホール輸送層、電子輸送性発光層、陰極である。
【０００４】上記素子では、ホール輸送層が、陽極から電子輸送性発光層へホールを注入する働きをするとともに、陰極から注入された電子がホールと再結合することなく陽極へ逃げるのを防ぎ、電子輸送性発光層内へ電子を封じ込める役割をも果たしている。このため、このホール輸送層による電子の封じ込め効果により、従来の単層構造の素子に比べてより効率よくホールと電子の再結合が起こり、駆動電圧の大幅な低下が可能となる。
【０００５】また斎藤らは、２層構造の素子において、電子輸送層だけでなくホール輸送層も発光層（ホール輸送性発光層）となり得ることを示した他〔C. Adachi, T. Tsutsui and S. Saito; Appl. Phys. Lett. , 55 (1989) 1489〕、ホール輸送層と電子輸送層の間に発光層が挟まれた３層構造の有機エレクトロルミネッセンス素子を提案した〔C. Adachi, S. Tokito, T. Tsutsui and S. Saito; Jpn. J. Appl. Phys. , 27 (1988) L269〕。
【０００６】斎藤らの２層構造の素子は、ガラス基板上に形成した陽極、ホール輸送性発光層、電子輸送層、陰極からなり、先のものと逆に、電子輸送層が、陰極からホール輸送性発光層へ電子を注入する働きをするとともに、陽極から注入されたホールが電子と再結合することなく陰極へ逃げるのを防ぎ、ホール輸送性発光層内へホールを封じ込める役割をも果たしている。このため、この電子輸送層によるホールの封じ込め効果により、先のものと同様に、駆動電圧の大幅な低下が可能となる。
【０００７】また斎藤らの３層構造の素子は、先のTangらの素子をさらに改良したもので、ガラス基板上に形成した陽極、ホール輸送層、発光層、電子輸送層、陰極からなり、ホール輸送層が電子を発光層に封じ込める働きをするとともに、電子輸送層がホールを発光層に封じ込める働きをするため、２層構造のものにくらべて、発光層内での電子とホールの再結合効率がさらに向上する。また上記電子輸送層、ホール輸送層は、電子とホールの再結合により生成した励起子が陰陽いずれかの電極層に逃げて消光されるのを防ぐ働きもする。したがって斎藤らの提案した３層構造の素子によれば、発光効率がさらに向上する。
【０００８】
【発明が解決しようとする課題】筒井によれば、エレクトロルミネッセンス素子の発光効率を示す外部エネルギー効率η_E (ext) は、素子の内部エネルギー効率η_E (int) および光の取り出し効率η_{EXT} と、式(a) ：
【０００９】
【数１】

$$\eta_E(ext) = \eta_{EXT}\,\eta_E(int) \qquad (a)$$

【００１０】の関係にある〔「有機ＥＬの最近の進歩」筒井哲夫、ディスプレィ　アンド　イメージング 1994, vol. 3, pp. 237-245〕。また上記のうち素子の内部エネルギー効率η_E (int) は、式(b) ：
【００１１】
【数２】

$$\eta_E(int) = \eta_\phi(int)\,\frac{\varepsilon_P}{V} \qquad (b)$$

【００１２】のごとく、内部量子効率$\eta\phi$(int) と、印加電圧Ｖと、放出されるフォトンの平均エネルギーε_P（ｅＶ）とによって求められ、さらに内部量子効率$\eta\phi$(int)は、式(c) ：
【００１３】
【数３】

$$\eta_{\phi}(\mathrm{int}) = \gamma\,\eta_r\,\eta_f \qquad (c)$$

【００１４】のごとく、電子とホールの注入バランス因子γと、キャリヤ再結合による一重項励起子の生成効率η_rと、一重項励起子からの発光量子効率η_fとによって求められる。そして従来、エレクトロルミネッセンス素子の発光効率の向上を目的として、素子の内部エネルギー効率η_E（int）を高めるべく、上記注入バランス因子γ、一重項励起子の生成効率η_rおよび一重項励起子からの発光量子効率η_fの向上について、種々検討がなされている。

【００１５】たとえば注入バランス因子γは、電子とホールが等量、注入される理想的な状態を１．０とする、電子とホールの注入バランスを示す係数であり、通常はγ＜１．０であるが、たとえば有機エレクトロルミネッセンス素子の場合は、前記のように有機の層を２層以上の多層構造として、電子とホールとの注入のバランスを改善することで、１．０に近づけることができる。

【００１６】また、電子とホールが再結合する際のスピン多重度の違いによって、三重項励起子と一重項励起子とが３：１の割合で発生するとの考えにしたがえば、一重項励起子の生成効率η_rの最大値は０．２５であるが、生成した三重項励起子が一重項励起子に転換することまでを考慮すると、上記生成効率η_rは０．４０まで増加する。

【００１７】さらに一重項励起子からの発光量子効率η_fは、主に蛍光色素等の発光物質固有の性質を反映しており、単純に孤立した分子１個からの発光を考慮すれば、発光物質分子、または分子固体に固有の蛍光量子効率：

【００１８】

【数４】

$$\Phi_{\phi}$$

【００１９】と等しいが、実際の素子においては、素子の層構成にともなう各種のエネルギープロセスによる一重項励エネルギーの損失がある。そこで、分子自体や分子固体自体の蛍光量子効率を大きくするとともに、前記のように有機の層を２層以上の多層構造とする等、素子の層構成を工夫して、一重項励起子からの発光量子効率η_fを１．０に近づけることが検討されている。

【００２０】一方、エレクトロルミネッセンス素子の発光効率を決定するもう一つの因子である光の取り出し効率η_{EXT}には、素子の光学的構造が係わっている。すなわち発光層での発光が大気中に放出されるには、幾つかの屈折率の異なる媒質の界面（素子の最外層と大気との界面を含む）を通過する必要があるが、スネルの屈折の法則にしたがえば、各界面に、その臨界角以上の角度で入射した光は、界面で全反射されて層中に封じ込められ、当該層中を導波して消失するので、その分だけ光の取り出し効率η_{EXT}が低下する。

【００２１】エレクトロルミネッセンス素子において、発光層を構成する屈折率ｎ（ｎ＞１．０）の媒体中で発生する面状発光を、屈折率が１．０である大気中に取り出す場合の、光の取り出し効率η_{EXT}は、上述した界面での全反射を考慮すると、上記屈折率ｎと、下記式(d)：

【００２２】

【数５】

$$\eta_{EXT} \sim \frac{1}{2n^2} \qquad (d)$$

【００２３】で表される関係にあることが知られている。これを有機エレクトロルミネッセンス素子に適用すると、発光層等の有機の層を構成する有機材料の屈折率ｎはおよそ１．６であるから、有機エレクトロルミネッセンス素子における光の取り出し効率η_{EXT}は約０．１９５となり、発光層で発生した光のおよそ２０％は素子外に取り出されるが、残り約８０％は、前記のように素子を構成する各層の界面での全反射により素子中に封じ込められ、層中を導波して消失することがわかる。

【００２４】このため、前述した内部エネルギー効率η_E（int）の向上と同時に、光の取り出し効率η_{EXT}を高めることが、今後の有機エレクトロルミネッセンス素子の実用化に向けて重要な課題である。たとえば猪口は、発光層で発光した光の、界面での全反射による層中への封じ込めを低減すべく、素子を構成する各層の界面を乱反射面としたり、あるいは発光層中に、散乱中心となる粒子を分散させることを提案している〔「エレクトロルミネッセントディスプレイ」猪口敏夫、産業図書株式会社刊、平成３年７月２５日初版発行〕。

【００２５】しかしこの提案の中で猪口は、上記の２法によって惹起される問題点についても指摘している。すなわち層の界面を乱反射面とする場合は、この乱反射面を構成する微小な凹凸のうち突起部に電界が集中するため、エレクトロルミネッセンス素子の駆動に高電圧が必要なことや、素子を構成する各層の膜厚が薄いこと等を考慮すると、上記突起部から、素子の絶縁破壊が生じるおそれがあるとしている。

【００２６】また発光層中に、散乱中心となる、明確に発達した粒界を有する粒子を分散させる場合は、当該粒子によって、発光層の電気的特性に局所的な不均一を生じ、それによって発光の不均一や絶縁破壊が生じるおそれがあるともしている。この発明の目的は、発光の不均一や絶縁破壊等を生じることなく、光の取り出し効率を向上させた、新規な有機エレクトロルミネッセンス素子を提供することにある。

【００２７】

【課題を解決するための手段】上記課題を解決するための、この発明の第１の有機エレクトロルミネッセンス素

子は、発光層を含む有機の層を、少なくとも一方が透明である一対の電極層で挟んだ構造の有機エレクトロルミネッセンス素子であって、少なくとも一方の電極層の、少なくとも有機の層との界面近傍、ならびに有機の層の、少なくとも一方の電極層との界面近傍のうち少なくとも一方に、２種以上の材料を不均一に分散して形成された、上記界面での光の反射・屈折角に乱れを生じさせる領域を設けたことを特徴としている。

【００２８】かかるこの発明の第１の有機エレクトロルミネッセンス素子においては、有機の層と電極層との界面の近傍に設けられた、上記界面での光の反射・屈折角に乱れを生じさせる領域により光が乱反射、乱屈折されるので、光の、全反射による層中への封じ込めが低減して、光の取り出し効率が向上する。また上記界面は平滑で、電界が集中するおそれのある凹凸を有しない上、この界面近傍に設けられた、当該界面での光の反射・屈折角に乱れを生じさせる領域は、２種以上の材料を分散したものであるが、両者は、層の電気的特性に局所的な不均一を生じるような明確に発達した粒界を有しない。

【００２９】よってこの発明の第１の有機エレクトロルミネッセンス素子によれば、発光の不均一や絶縁破壊等を生じることなく、光の取り出し効率を向上させることが可能となる。また、この発明の第２の有機エレクトロルミネッセンス素子は、発光層を含む有機の層を備えた有機エレクトロルミネッセンス素子であって、当該素子のうち、光の取り出し側の表面を乱反射面とするか、または上記光の取り出し側の表面近傍に、２種以上の材料を不均一に分散して形成された、上記表面での光の反射・屈折角に乱れを生じさせる領域を設けたことを特徴とするものである。

【００３０】上記この発明の第２の有機エレクトロルミネッセンス素子は、発光層の電気的特性に関係しない素子の光の取り出し側の表面を、前記のように乱反射面とするか、または上記表面近傍に、当該表面での光の反射・屈折角に乱れを生じさせる領域を設けたものゆえ、発光の不均一や絶縁破壊等を生じることなく、光の、全反射による層中への封じ込めを低減して、光の取り出し効率を向上させることができる。

【００３１】なお、ここでいう光の反射・屈折角に乱れを生じさせる領域とは、有機の層のうち発光層で発光し、電極層との界面、または素子の光の取り出し側の表面に入射した光の、当該界面または表面での反射角度および屈折角度のうちの一方または両方に乱れを生じさせる機能を有する領域である。

【００３２】

【発明の実施の形態】まず、この発明の第１の有機エレクトロルミネッセンス素子について説明する。上記第１の有機エレクトロルミネッセンス素子は、少なくとも発光層を含む有機の層を、少なくとも一方が透明である一対の電極層で挟んだ構造を有するものである。

【００３３】有機の層は、従来どおり、発光層のみを有する単層構造であってもよく、また発光層を、前述したように種々の層と組み合わせた２層あるいは３層、４層等の多層構造であってもよい。２層構造の例としては、たとえば前述したホール輸送性発光層と電子輸送層との組み合わせや、あるいは電子輸送性発光層とホール輸送層との組み合わせ等があげられる。

【００３４】また３層構造の例としては、たとえば前述したホール輸送層と発光層と電子輸送層との組み合わせの他、ホール輸送性発光層と、陽極から注入されたホールが電子と再結合することなく陰極へ逃げるのを防ぎ、ホール輸送性発光層内へホールを封じ込める働きをするホールブロック層と、電子輸送層との組み合わせや、あるいは電子輸送性発光層と、陰極から注入された電子がホールと再結合することなく陽極へ逃げるのを防ぎ、電子輸送性発光層内へ電子を封じ込める働きをする電子ブロック層と、ホール輸送層との組み合わせ等があげられる。

【００３５】さらに４層構造の例としては、たとえば陽極からホール輸送層にホールが注入されるのを助ける働きをするホール注入層と、ホール輸送層と、発光層と、電子輸送層との組み合わせや、あるいは上記ホール注入層と、ホール輸送性発光層と、ホールブロック層と、電子輸送層との組み合わせ等があげられる。上記有機の層は、当該有機の層に電子およびホールを注入するための陰陽両電極層と組み合わされる。この際、発光層からの発光を素子外に取り出すために、上記両電極層のうちの少なくとも一方は、前述したように、ＩＴＯ（インジウムチンオキサイド）等の透明導電材料にて形成される。

【００３６】素子の保護を考慮すると、上記各層が積層、形成される基材を、ガラスや透明プラスチックフィルム等で形成して、当該基材側を、発光層からの光の取り出し側とするのが好ましく、この場合には、基材の直上に形成される側の電極層を、上記透明導電材料にて形成すればよい。また、有機の層の上に形成される側の電極層の上には、素子を空気中の水分や酸素等から保護すべく、保護層を形成したり、あるいは保護層を形成した素子を、さらにガラスやポリマーで封止してもよい。

【００３７】この発明においては、上記各層構造の有機エレクトロルミネッセンス素子のうち、前述したように少なくとも一方の電極層の、少なくとも有機の層との界面近傍、および／または上記有機の層の、少なくとも一方の電極層との界面近傍に、２種以上の材料を不均一に分散した、光の反射・屈折角に乱れを生じさせる領域が設けられる。

【００３８】具体的には、(1) 有機の層を挟む一対の電極層の一方または両方の、有機の層との界面近傍、または電極層の全体、(2) 有機の層のうち、一対の電極層の一方または両方との界面近傍、(3) 上記(1)(2)の両方、のいずれかに、光の反射・屈折角に乱れを生じさせる領

域が形成される。

【００３９】上記領域を所定の箇所に設けるには、種々の方法が考えられるが、たとえば真空蒸着法等の、気相成長法により形成される層に、上記の領域を設けるには、当該層を、互いに異なる２種以上の材料の同時蒸着により形成したり、あるいは一方の材料で形成した層に、他方の材料をドーピングしたりすればよい。たとえば透明でない側の電極層に、光の反射・屈折角に乱れを生じさせる領域、より詳しくは光の反射角に乱れを生じさせる領域を設けるには、当該電極層を、後述する実施例のごとくマグネシウム（Ｍｇ）と銀（Ａｇ）との、蒸着速度比１０：１程度の共蒸着により形成すればよい。

【００４０】このようにして形成された電極層は、Ｍｇの連続相中に、ＭｇＡｇ合金相が明確な粒界を有しないで分散された構造となり、上記のように発光層からの光の反射角に乱れを生じる結果、光の、全反射による層中への封じ込めを低減して、光の取り出し効率を向上させる。なお上記電極層において、発光層からの光の反射角に乱れを生じる原因としては、たとえば以下の２つが考えられる。

①　電極層の、有機の層との界面は、Ｍｇ相、ＭｇＡｇ相、および両者の中間相（前記のように両相が明確に発達した粒界を有しないため）等の様々な相になっており、それぞれの相の、入射光に対する相互作用が異なるために反射角に差が生じ、その結果、発光層からの光の反射角に乱れを生じる。

②　素子作製の際に雰囲気中、あるいは有機の層中に含まれる酸素により、上記電極層と有機の層との界面がごくわずかに酸化されており、その酸化層の組成が、電極層を構成する上記各相によって異なるため、光の屈折率にも違いが生じ、その結果、発光層からの光の反射角に乱れを生じる。

【００４１】なお、上記電極層以外の透明な層に、光の反射・屈折角に乱れを生じる領域を形成するには、たとえば２種以上の屈折率の異なる透明材料を不均一に分散すればよい。光の反射・屈折角に乱れを生じさせる、２種以上の材料の分散構造において、上記分散構造の大きさ（たとえば上記Ｍｇの連続相中に分散されたＭｇＡｇ合金相の大きさ等）はとくに限定されないが、発光層からの発光波長の１／１０～１０倍程度であるのが好ましい。

【００４２】分散構造の大きさが上記範囲未満であったり、あるいは上記範囲を超えた場合には、反射・屈折角の乱れにより、発光層からの発光の、界面での全反射を低減させる効果が不十分になるおそれがある。なお上記分散構造の大きさは、上記範囲内でもとくに、発光波長と等倍以下であるのがより一層、好ましい。

【００４３】つぎに、この発明の第２の有機エレクトロルミネッセンス素子について説明する。上記第２の有機エレクトロルミネッセンス素子は、発光層を含む有機の層を備えるものである。有機の層の層構成は、先の第１の有機エレクトロルミネッセンス素子の場合と同様に単層構造でも、あるいは２層以上の積層構造でもよい。

【００４４】電極層ならびに基材についても、先の第１の有機エレクトロルミネッセンス素子の場合と同様に構成できる。すなわち、素子の保護を考慮すると、有機の層および電極層が積層、形成される基材を、ガラスや透明プラスチックフィルム等の透明基材として、当該基材側を、発光層からの光の取り出し側とするのが好ましく、この場合には、基材の直上に形成される側の電極層を、透明導電材料にて形成すればよい。

【００４５】また、有機の層の上に形成される側の電極層の上には、素子を空気中の水分や酸素等から保護すべく、保護層を形成したり、あるいは保護層を形成した素子を、さらにガラスやポリマーで封止してもよい。この発明においては、上記各層構造の有機エレクトロルミネッセンス素子のうち、前述したように光の取り出し側の表面を乱反射面とするか、または上記光の取り出し側の表面近傍に、２種以上の材料を不均一に分散して形成された、光の反射・屈折角に乱れを生じさせる領域が設けられる。

【００４６】具体的には、

①　光の取り出し側が、前記のように透明基材側である場合に、当該透明基材の、各層が形成される側と反対側の表面、

②　光の取り出し側が、基材と反対側の電極層側である場合に、当該電極層の表面、

③　上記電極層の表面に保護層が形成される場合に、当該保護層の表面、のいずれかが乱反射面とされるか、または上記各表面の近傍に、光の反射・屈折角に乱れを生じさせる領域が形成される。

【００４７】なおこのうち①の透明基材は、その表面を機械研磨等の方法で粗化して乱反射面とするのが、作業性等の点で好ましい。一方、②の電極層や③の保護層は主として蒸着法で形成されるものゆえ、前記のように互いに異なる２種以上の材料の同時蒸着により形成したり、あるいは一方の材料で形成した層に、他方の材料をドーピングしたりすることで、表面近傍に、光の反射・屈折角に乱れを生じさせる領域を形成するのが好ましい。

【００４８】上記乱反射面を構成する凹凸の大きさはとくに限定されないが、乱反射面の凹凸の程度を表すＲＭＳ値が、発光層からの発光波長の１／１０～１０倍程度であるのが好ましい。ＲＭＳ値が上記範囲未満であったり、あるいは上記範囲を超えた場合には、乱反射により、発光層からの発光の、界面での全反射を低減させる効果が不十分になるおそれがある。

【００４９】なお上記ＲＭＳ値は、上記範囲内でもとくに、発光波長と等倍以下であるのがより一層、好ましい。一方、光の反射屈折角に乱れを生じさせる領域を構

成する、２種以上の材料の分散構造の大きさは、先の第１の有機エレクトロルミネッセンス素子の説明で述べたのと同範囲であるのが好ましい。

【００５０】上記第２の有機エレクトロルミネッセンス素子においては、さらに、有機の層と、それを挟む一対の層のうち少なくとも一方との界面に、前述した第１の有機エレクトロルミネッセンス素子の構成を導入してもよい。すなわち上記界面近傍に、２種以上の材料を不均一に分散して形成された、光の反射・屈折角に乱れを生じさせる領域を設けてもよい。この場合には、光の取り出し側の表面における全反射の低減効果と、上記界面における全反射の低減効果とが相まって、素子の発光効率をさらに向上することができる。

【００５１】

【実施例】以下にこの発明を、実施例、比較例に基づいて説明する。

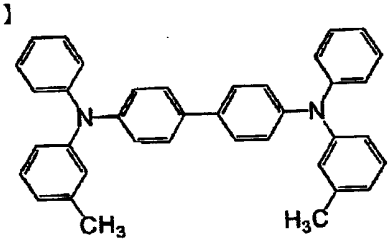
実施例１

電子ビーム蒸着法にてＩＴＯ（インジウムチンオキサイド）を蒸着して、厚み１５０～１６０ｎｍ、シート抵抗１５Ω／□のＩＴＯ電極層を形成したガラス基材を、洗剤（セミコクリーン）、蒸留水、アセトン、およびイソプロパノールをこの順に用いて超音波洗浄した後、煮沸メタノール中で洗浄、乾燥させ、さらにＵＶオゾンアッシングにて清浄化した。

【００５２】つぎにこのガラス基材を真空蒸着装置にセットし、発光領域の形状に対応するマスクを装着した状態で、到達真空度：1×10^{-6}Ｔｏｒｒ、基板温度：室温、蒸着速度０．２～０．３ｎｍ／ｓの条件下、真空蒸着法によって、上記ＩＴＯ電極層上に、まず式(1)：

【００５３】

【化１】

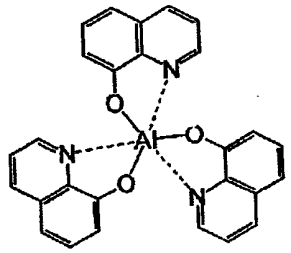


(1)

【００５４】で表されるＮ，Ｎ′－ジフェニル－Ｎ，Ｎ′－ビス（３－メチルフェニル）－１，１′－ビフェニル－４，４－ジアミンを蒸着して、膜厚４０ｎｍのホール輸送層を形成し、つぎにこのホール輸送層上に、引き続き同条件で、真空蒸着法によって、式(2)：

【００５５】

【化２】



(2)

【００５６】で表されるトリス（８－キノリノラート）アルミニウム(III) 錯体を蒸着して、膜厚６０ｎｍの電子輸送性発光層を形成した。つぎに大気中で、マスクを陰極の形状に対応するものと交換した後、到達真空度：1×10^{-6}Ｔｏｒｒ、基板温度：室温、蒸着速度１ｎｍ／ｓの条件下、真空蒸着法によって、上記電子輸送層上にＭｇとＡｇを蒸着速度比１０：１で共蒸着して、Ｍｇ／Ａｇ＝１０／１（モル比）、膜厚１５０ｎｍのＭｇ／Ａｇ電極層を形成した後、その上にＡｇを単独蒸着して、膜厚１００ｎｍの保護層を形成して、有機エレクトロルミネッセンス素子を製造した。

【００５７】そして、上記有機エレクトロルミネッセンス素子のＩＴＯ電極層を陽極、Ｍｇ／Ａｇ電極層を陰極として室温、大気中で両電極間に直流電圧を印加したところ、４Ｖ以上の電圧で、電子輸送性発光層からの緑色発光（発光ピーク５３ｎｍ）が観察された。また上記素子の発光輝度を、輝度計〔ミノルタ（株）製のＬＳ－１００〕を用いて測定したところ、最高輝度は２００００ｃｄ／m^2 であり、またこの結果から素子の発光効率を求めたところ、最高発光効率は１．５１ｍ／Ｗであった。

【００５８】さらに、上記Ｍｇ／Ａｇ電極層をＸ線回折測定したところ、かかるＭｇ／Ａｇ電極層は、図１に示すようにＭｇのピークに加えてＭｇＡｇ合金のピークが検出されたことから、Ｍｇの連続相中に、ＭｇＡｇ合金相が分散された構造を有することがわかった。そして上記Ｍｇ／Ａｇ電極層の、金属顕微鏡による観察から、上記ＭｇＡｇ合金相は、Ｍｇの連続相中に、明確な粒界を有しない状態で分散されていることがわかった。

比較例１

Ｍｇ／Ａｇ電極層に代えて、Ｍｇの単独蒸着層（膜厚１５０ｎｍ）を形成したこと以外は、実施例１と同様にして有機エレクトロルミネッセンス素子を製造した。

【００５９】そして、上記有機エレクトロルミネッセンス素子のＩＴＯ電極層を陽極、Ｍｇ／Ａｇ電極層を陰極として室温、大気中で両電極間に直流電圧を印加したところ、４Ｖ以上の電圧で、電子輸送性発光層からの緑色発光（発光ピーク５３ｎｍ）が観察された。また上記素子の発光輝度を、輝度計〔ミノルタ（株）製のＬＳ－１００〕を用いて測定したところ、最高輝度は８０００ｃｄ／m^2 であり、またこの結果から素子の発光効率を求

めたところ、最高発光効率は１．０１ｍ／Ｗであった。

実施例２

素子の光の取り出し側の表面に相当する、ガラス基材の、ＩＴＯ電極層を形成する側と反対側の表面を、表面粗度がＲＭＳ値で１０～２０ｎｍとなるように機械研磨したこと以外は実施例１と同様にして有機エレクトロルミネッセンス素子を製造した。

【００６０】そして、上記有機エレクトロルミネッセンス素子のＩＴＯ電極層を陽極、Ｍｇ／Ａｇ電極層を陰極として室温、大気中で両電極間に直流電圧を印加したところ、４Ｖ以上の電圧で、電子輸送性発光層からの緑色発光（発光ピーク５３ｎｍ）が観察された。また上記素子の発光輝度を、輝度計〔ミノルタ（株）製のＬＳ－１００〕を用いて測定したところ、最高輝度は４３０００ｃｄ／m^2 であり、またこの結果から素子の発光効率を求めたところ、最高発光効率は３．８１ｍ／Ｗであった。

実施例３

Ｍｇ／Ａｇ電極層に代えて、蒸発物質としてアルミニウム（Ａｌ）リチウム（Ｌｉ）合金（Ｌｉ農度２重量％）を用いた真空蒸着法により、膜厚１５０ｎｍ、Ｌｉ農度０．０５重量％のＡｌ／Ｌｉ電極層を形成したこと以外は、実施例２と同様にして有機エレクトロルミネッセンス素子を製造した。

【００６１】そして、上記有機エレクトロルミネッセンス素子のＩＴＯ電極層を陽極、Ａｌ／Ｌｉ電極層を陰極として室温、大気中で両電極間に直流電圧を印加したところ、４Ｖ以上の電圧で、電子輸送性発光層からの緑色発光（発光ピーク５３ｎｍ）が観察された。また上記素子の発光輝度を、輝度計〔ミノルタ（株）製のＬＳ－１００〕を用いて測定したところ、最高輝度は２１０００ｃｄ／m^2 であり、またこの結果から素子の発光効率を求めたところ、最高発光効率は１．６１ｍ／Ｗであった。

【００６２】なお上記Ａｌ／Ｌｉ電極層を金属顕微鏡により観察したところ、ＡｌＬｉ合金の均一な連続層であることがわかった。

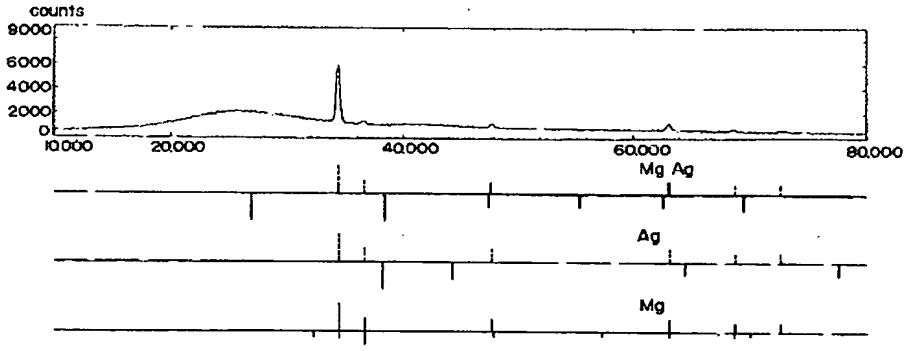
【００６３】

【発明の効果】以上、詳述したようにこの発明の有機エレクトロルミネッセンス素子は、有機の層と電極層との界面近傍、または素子の光の取り出し側の表面近傍に、２種以上の材料を不均一に分散して形成された、光の反射・屈折角に乱れを生じさせる領域を設けるか、または素子の光の取り出し側の表面を乱反射面としたものゆえ、発光の不均一や絶縁破壊等を生じることなく、光の取り出し効率を向上させることが可能となる。

【図面の簡単な説明】

【図１】この発明の、実施例１で製造した有機エレクトロルミネッセンス素子における、ＭｇＡｇ電極層のＸ線回折スペクトルを示すグラフである。

【図１】



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